

Graphene, the Wonder -material

"A suspended sheet of pure Graphene – a plane layer of Carbon atoms bonded together in a honeycomb lattice – is the "most two-dimensional" system imaginable."

A.J. Leggett, Nobel Laureate in Physics

Abstract

Graphene has attracted a great interest in material science due to its novel electronic structures. Owing to this unusual electronic spectrum, Graphene has led to the emergence of a new branch of 'relativistic' condensed-matter physics, where quantum relativistic phenomena, some of which are unobservable in high-energy physics, can be simulated and tested in the laboratories. More generally, Graphene represents a conceptually new class of materials that are only one atom thick and offers new opportunities into low-dimensional physics. Magnetism discovered in Graphene based systems opens up the possibility of their spintronics application. Theoretical and experimental studies suggest that such magnetism in Graphene mainly comes from the localized states or edges states. This paper provides an outline on the electronic structures and magnetic behaviors of 2-dimensional Graphene and its possible applications.

Introduction

Graphene is a single layer of carbon packed in a hexagonal (honeycomb) lattice. First discovered in 2004 by Novoselov et al., the single atomic layer of Graphene has a thickness of only 0.34 nm of sp^2 hybridized carbon atoms covalently bonded to three other atoms arranged in a honeycomb pattern with a carbon-carbon distance of 0.142 nm. It is the first truly two-dimensional crystalline material and is representative of a whole class of 2D materials. Graphene forms the basic structure of other carbon-based materials such as fullerene (wrapped-up Graphene), carbon nanotubes (several Graphene sheets rolled up along a vertical axis) and graphite (stacked Graphene).

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Graphene's unique structural (truly 2 dimensional), mechanical (high tensile strength), optical (Graphene is

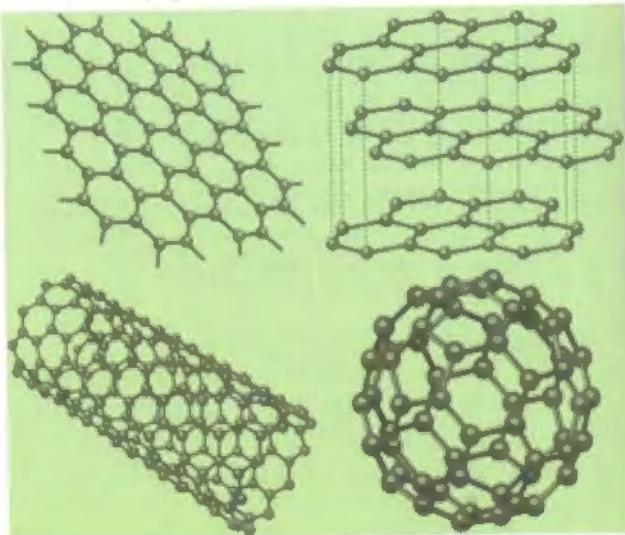


Figure 1: From Graphene to Bucky ball, Carbon Nanotubes and Graphite

nearly transparent), thermal (its thermal conductivity is much higher than that of silver), electrical (particularly high carrier mobility and Anomalous Quantum Hall Effect at room temperature) and magnetic properties make it one of the most important topics in materials science today. Graphene, due to its strange properties, appears to have tremendous potential applications as Nanoelectronic devices, chemical sensors, hydrogen storage systems or polymer Nano-composites. Graphene could also be considered as a prototypical material to study the properties of other two-dimensional Nanosystems such as single layers of Boron nitride, Molybdenum disulphide, Silicon carbide, Silicon, Germanium and Zinc oxide.

The Discovery of Graphene

Graphene had already been studied theoretically in 1947 by Canadian physicist P.R. Wallace as a text book example for calculations in solid state physics. He predicted the electronic structure and noted the linear dispersion relation. He also predicted the relativistic behavior of electrons in Graphene. The wave equation for excitations was written down by J.W. McClure *already in 1956, and the similarity to the* Dirac equation was discussed by G.W. Semenoff in 1984. It took another twenty years for the discovery of Graphene in the laboratory when Andre Geim, Konstantin Novoselov and their collaborators from the University of Manchester (UK), and the Institute for Microelectronics Technology in Chernogolovka (Russia), presented their results on Graphene structures in October 2004 issue of *Science*.



Figure 2: Andre Geim (left) with Konstantin Novoselov

In this paper Geim and Novoselov described the fabrication, identification and characterization of

Graphene by Atomic Force Microscopy (AFM). They used a simple mechanical exfoliation method for extracting thin layers of graphite from a graphite crystal with Scotch tape and then transferred these layers to a silicon substrate. This method was first suggested and tried by R. Ruoff's group who were, however, not able to identify any monolayers. The Manchester group succeeded by using an optical method with which they were able to identify fragments made up of only a few layers. In some cases these flakes were made up of a single layer, i.e. Graphene was identified. Moreover, they managed to make Hall measurements of Graphene.

In December 2004, just two months after the paper by Novoselov et al. had been published, a group led by W. A. de Heer at Georgia Tech published their first paper on transport measurements on thin carbon films. They presented magneto-resistance measurements and also a weak electric field effect

A group at the University of Columbia led by P. Kim investigated an alternative approach for making thin carbon layers. They attached a graphite crystal to the tip of an atomic force microscope and dragged it along a surface. In this way they were able to produce thin layers of graphite down to approximately 10 layers.

Since the first successful isolation of Graphene in the laboratory, it has attracted tremendous attention within the scientific community. This is mainly due to the fact that Graphene is an almost perfect two-dimensional system with unique electronic properties that might ultimately lead to a new generation of Nano electronic devices.



Figure 3: Graphene pellets and flakes



The Electronic Properties of Graphene

Graphene's strange behavior arises due to its peculiar electronic configuration. Graphene's stability is due to a tightly packed, periodic array of carbon atoms and a sp^2 orbital hybridization - a combination of orbitals p_x and p_y that constitute the σ -bond. Graphene has three σ -bonds and one π -bond. The final p_z electron makes up the π -bond, and is key to the half-filled band that permits free-moving electrons. Graphene is practically transparent in the optical region as it absorbs only 2.3% of visible light. Mechanical studies of Graphene have shown that it is mechanically extremely strong, a hundred times stronger than the strongest steel, very stretchable and can be used as a flexible conductor. Graphene is the strongest material ever tested, with an intrinsic Tensile strength of 130 GPa and a Young's modulus of 1 TPa. Its thermal conductivity is much higher than that of silver. The lattice contribution of Specific Heat of Graphene varies as T^3 at low temperature (T) while at

higher temperature it varies as $T^{3/2}$, quite unlike Graphite whose Specific Heat varies as T^2 in the lower temperature but at the higher range, it is nearly constant. In contrast to low temperature 2D systems based on semiconductors, Graphene maintains its 2D properties even at room temperature. Graphene also has several other interesting properties, which it shares with carbon nanotubes. A truly unique feature is that Graphene is structurally malleable and its electronic, optical and phonon properties can be strongly modified by strain and deformation.

The real surprise is the electronic properties of Graphene. As a result, the most explored aspect of Graphene physics is also its electronic properties. Graphene's electronic properties are truly unique and are different from those of any other known condensed matter system. These unique properties arise from the collective behavior of electrons. We know that when a large number of particles interact strongly with each

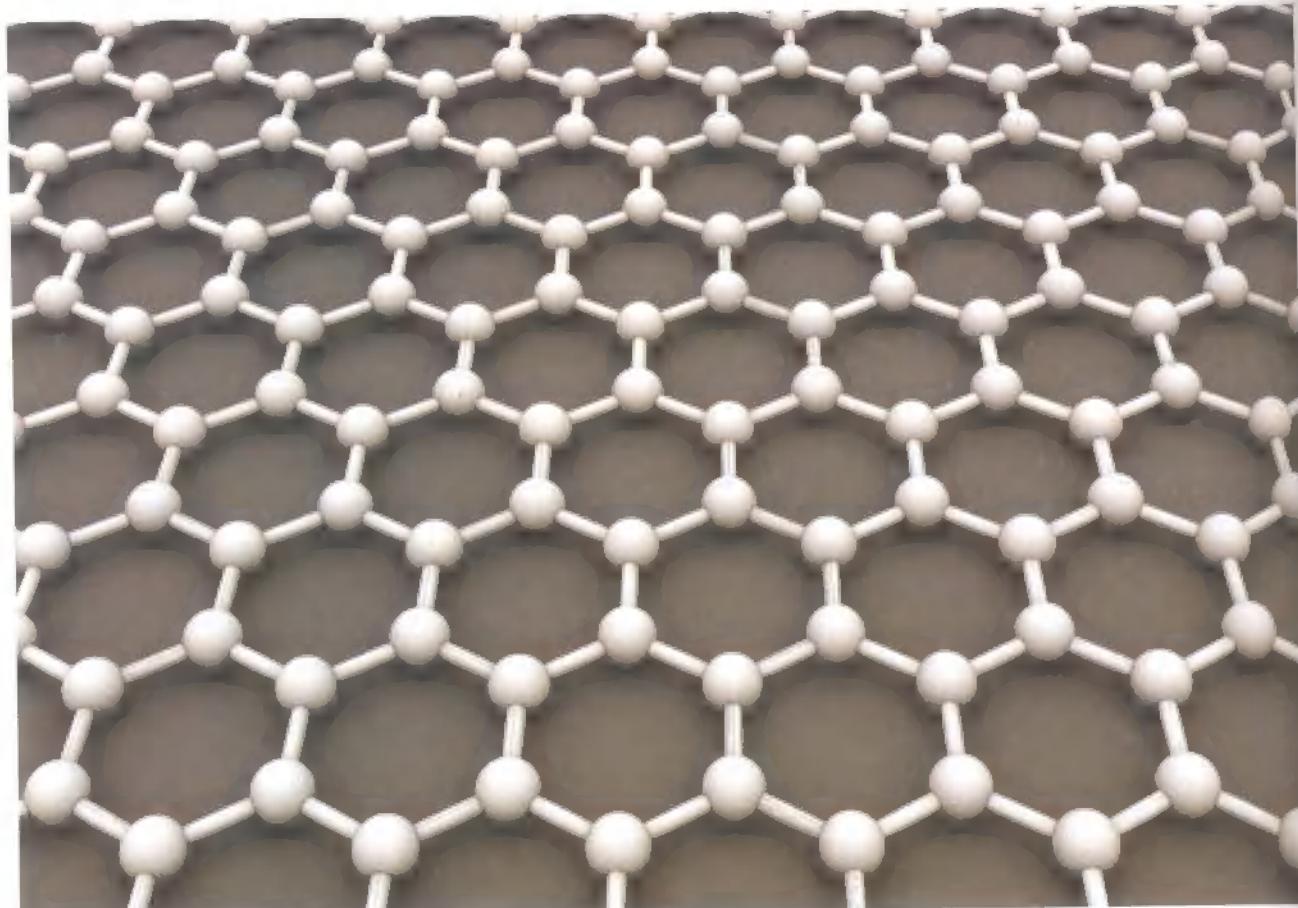


Figure 4a. A model of Graphene

other, unexpected collective motions can emerge. In the case of Graphene, however, the interaction between electrons and the honeycomb lattice causes the electrons to behave as if they have absolutely no mass. Electron waves propagating through the honeycomb lattice completely lose their effective mass resulting in quasiparticle with zero rest mass.

In any crystal, the ordered arrangement of atoms creates a periodic potential, which causes electrons to move in waves called Bloch waves. Just as we associate photons with light waves, we can associate each Bloch wave with a "quasiparticle" that has an energy $E = \hbar\omega$ (where \hbar is the Reduced Planck's constant and ω is the angular frequency of the wave). Strictly speaking, these quasiparticles are not electrons, but physicists wrongly give them this name. The electrons or the quasiparticles move in this periodic potential with a mass called effective mass (m^*) which is different from electronic mass. Generally electrons in solids move with velocity much slower than the velocity of light and hence relativistic corrections are not required. But electrons in Graphene move at an effective speed of light which is only 300 times less than the speed of light in a vacuum, allowing relativistic effects to be observed without using particle accelerators. This relativistic velocity of electrons in Graphene allows scientists to use and test Relativistic Quantum Mechanics or Dirac formalism rather than Schrödinger formalism for Graphene. Strangely, only a honeycomb lattice produces this peculiar property; atoms arranged in other periodic structures, such as square or triangular lattices, always generate electrons with a finite effective mass.

Charge carriers in crystalline solid are normally described by the Schrödinger equation with an effective mass m^* different from the free electrons. Relativistic particles in the limit of zero rest mass follow the Dirac equation. Charge carriers in Graphene are, therefore, called massless Dirac fermions and described by a 2D analogue of the Dirac equation with the Fermi velocity $V_F \approx 1 \times 10^6$ m/s playing the role of the speed of light and a 2D pseudo spin matrix σ describing two sub lattices of the honeycomb lattice. Similar to the real spin that can change its direction between, say, up and down, the pseudo spin is an index that indicates on which of the two sub lattices a quasiparticle is located.

The formal similarity between the excitations in Graphene and two-dimensional Dirac fermions has

allowed testing of the so called Klein tunneling which was suggested by the Swedish physicist Oskar Klein. Klein paradox predicts that a very large potential barrier becomes completely transparent to relativistic electrons as opposed to calculations using Schrödinger's equation for a nonrelativistic particle. Probability that an electron "tunnels" through a potential barrier falls exponentially with the height of the barrier. However, calculations show that for relativistic particles the tunnelling probability increases with the barrier height, since a potential barrier that repels electrons will also attract their antiparticles. This effect has been displayed much more easily with the massless Dirac fermions in Graphene, as already suggested by Geim and coworkers. The existence of antiparticles is a result of Relativistic Quantum Mechanics, as 'holes' exist in semiconductors as antiparticles of electrons.

Another important observation is that the electron mobility of Graphene is about ten times higher than the mobility of commercial silicon wafers and electrons can travel huge distances (300 nm or more) without being scattered. Graphene's quality clearly reveals itself in a pronounced ambipolar electric field effect such that charge carriers can be tuned continuously between electrons and holes in concentrations n as high as 10^{13} cm^{-2} and their mobilities μ can exceed $15,000 \text{ cm}^2/\text{Vs}$ even under ambient conditions. Moreover, the observed mobilities weakly depend on temperature T , which means that μ at 300K is still limited by impurity scattering and, therefore, can be improved significantly, perhaps, even up to $\approx 100,000 \text{ cm}^2/\text{Vs}$. Although some semiconductors exhibit room-temperature μ as high as $\approx 77,000 \text{ cm}^2/\text{Vs}$ (namely, InSb), those values are quoted for undoped bulk semiconductors. In Graphene, μ remains high even at high $n (> 10^{13} \text{ cm}^{-2})$ in both electrically and chemically-doped devices, which translates into ballistic transport on submicron scale (up to $\approx 0.3 \mu\text{m}$ at 300K). These excellent properties make Graphene a potential substitute for silicon in electronics. Electrons in Graphene can cover submicron distance without scattering, even in samples placed on an atomically rough substrate and at room temperature. Due to the massless carriers and little scattering, quantum effects in Graphene are very prominent and can be observed even at room temperature.

Electron waves in Graphene propagate within a layer that is only one atom thick, which makes them sensitive to the proximity of other materials such as high-k

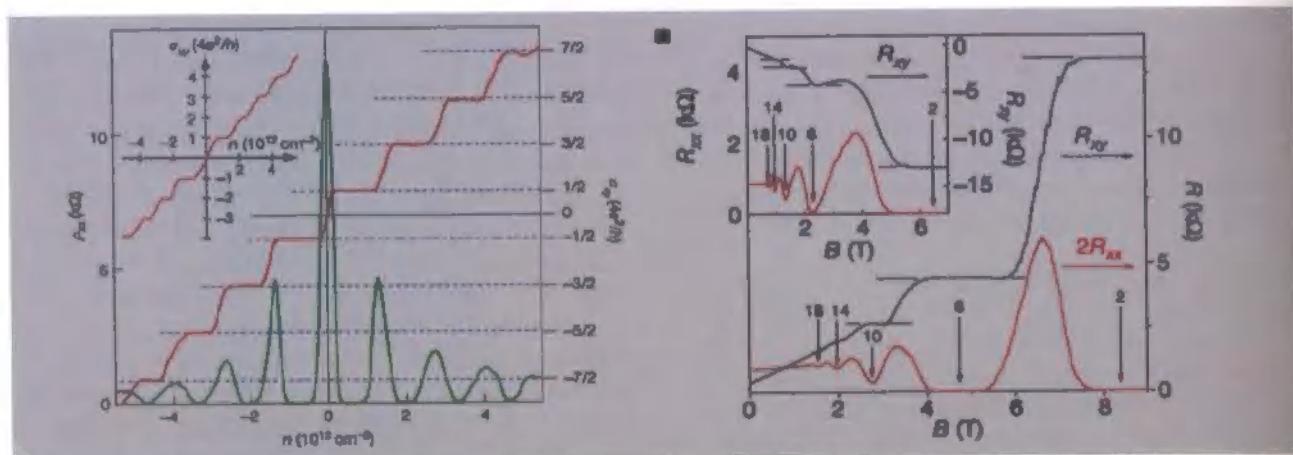


Figure 5: Anomalous Integral Quantum Hall Effect in Graphene

dielectrics, superconductors, ferromagnetic materials etc. This feature offers many enticing possibilities in comparison with the conventional 2D electronic systems (2DES).

The electrical conductivity of intrinsic Graphene is quite low and is of the order of the conductance quantum $\sigma \sim e^2/h$. The Fermi level can however be changed by an electric field so that the material becomes either n-doped (with electrons) or p-doped (with holes) depending on the polarity of the applied field. Graphene can also be doped by adsorbing, for example, water or ammonia on its surface. The electrical conductivity for doped Graphene is potentially quite high; at room temperature it may even be higher than that of copper.

Hall Effect is a galvano-magnetic phenomenon where transverse conductivity can be observed in the presence of a strong magnetic field. For metals and bulk semiconductors at room temperature, the relation between transverse conductivity and applied magnetic field is linear. In 1980 Klaus von Klitzing discovered the Quantum Hall Effect (QHE) which tells us that that in a 2D electron gas at a temperature close to absolute zero the Hall resistivity becomes quantized, taking only discrete values of h/ne^2 (where h is Planck's constant, n is a positive integer and e is the electric charge). The quantization is so precise that this Quantum Hall Effect is used as the standard for the measurement of resistivity. The quantization of the Hall Effect at integer multiples of the basic quantity h/ne^2 can usually be observed only in very thin and pure silicon or gallium arsenide solids at temperatures around ≤ 3 K and very high magnetic fields. Graphene shows the quantum Hall

effect with respect to conductivity-quantization: the effect is anomalous in that the sequence of steps is shifted by 1/2 with respect to the standard sequence and with an additional factor of 4. These anomalies are present at room temperature, i.e. at roughly 20 °C. This behavior is a direct result of Graphene's massless Dirac electrons. In a magnetic field, their spectrum has a Landau level with energy precisely at the Dirac point.

Graphene opens up many possibilities in Particle Physics also. Neutrinos are massless Dirac fermions in high-energy particle physics like charge carriers in Graphene. But neutrinos have no electric charge and therefore do not interact strongly with any kind of matter. The Dirac Fermions in Graphene, in contrast, carry one unit of electric charge and so can be manipulated using electromagnetic fields. Since the manipulation of electrons within materials is at the heart of modern electronics, the radically different behavior of electrons in Graphene may allow us to go beyond the limits of silicon-based semiconductor technology.

Graphene may also help address the puzzle of "chiral symmetry breaking". The chirality of a particle tells us whether it differs from its own mirror image, like a right-handed and left-handed screw, for example. In Graphene there are "left-handed" and "right-handed" Dirac fermions, but they behave in the same way as each other. This is in stark contrast to neutrinos, which only appear in their left-handed form. Whether or not the symmetry between the left-handed and right-handed particles in Graphene can be broken may help us understand how the same symmetry is broken in particle physics.

Another very interesting property of Graphene is the modification of Fine Structure constant (α). In Quantum Electrodynamics (QED), the strength of electromagnetic interactions between charged particles is described by the fine-structure constant, $\alpha = e^2/hc$, where h is Planck's constant divided by 2π and c is the speed of light. With a value of 1 divided by $137.03599911 \pm 0.00000046$, this is one of the most precisely measured physical quantities in nature. This is also the ratio of the speed of the revolving electron in the first hydrogen orbit and the velocity of light. Since the effective speed of light for the Dirac fermions in Graphene is 300 times less, Graphene's fine-structure constant should have a much larger value of about two. This example shows that physical constants like α can be modified by the presence of a complex environment, in this case Graphene's honeycomb lattice. Why is this arbitrary value of the fine-structure constant after all? Perhaps the answer has been given by Xiao-Gang Wen of the Massachusetts Institute of Technology, that the electron is not as elementary as one would think, but is instead a consequence of interactions between more complex degrees of freedom not yet experimentally accessible.

Magnetism in Graphene

Magnetism in Graphene is an emerging field that has received much theoretical attention. Graphene, as a metal-free material, contains no magnetic atoms. Graphene does not have any d and f shell electrons which are responsible for the magnetic coupling in conventional Ferro magnets. Its honeycomb structure contains a bipartite lattice, formed by two interpenetrating triangular sub-lattices (A and B). The electronic structure of Graphene can be described by a single orbital nearest-neighbor hopping Hamiltonian. This model correctly describes the Graphene with linear bands around the Fermi energy. Despite this fact, both theory and experiments suggest that a magnetic order can exist in these carbon structures under particular circumstances. The magnetism in Graphene comes from the local states caused by defects or molecular adsorption.

There have been exciting predictions for induced magnetism through proximity to a ferromagnetic insulator as well as through localized dopants and defects. Ferromagnetism has been shown to exist in Graphene subjected to irradiation. Glassy carbon produced by laser ablation is also reported to exhibit

ferromagnetic-like behavior with magnetic hysteresis. Localized unpaired spins arising from topological and bonding defects are suggested to be responsible for ferromagnetism observed in such nanomaterial.

Defects in ideal Graphene can be introduced by both vacancies and external doping. Many experimental works have reported the existence of magnetism in carbon materials by electrons or ions irradiation. The common feature of these defects is that carbon atoms are removed from the Graphene sheet, which gives quasilocalized states at the Fermi level. In this scenario, adsorbates or lattice vacancies effectively remove a p_z -orbital from the Graphene band structure. This is known to create a localized defect state near the Fermi energy. Due to the Coulomb interaction, the defect state is spin split leading to a spin-1/2 populated quasilocalized defect state. Whereas bulk Graphene is a diamagnetic semimetal, simple tight-binding models predict that one-dimensional ribbons with zigzag edges are paramagnetic metals. We know that Graphene has a bipartite lattice structure. The two sub lattices have different spin populations. It is argued that only when these defects are produced in the same sub lattice, the magnetic moments from the defective states can make a ferromagnetic (FM) coupling. Besides the native defects (vacancies), Graphene can also show magnetism by doping defects. Studies found that additional hydrogen adsorption on the vacancy can destroy the magnetism. On the other hand, the presence of nitrogen or Boron around a vacancy can produce larger macroscopic magnetic signals as compared to a standalone carbon vacancy. The π orbitals of the atoms around the border regions of graphite and BN are localized, and they are responsible for the magnetism. Theoretical calculations reveal that such magnetic atoms favor Ferromagnetic coupling. The different electronic and magnetic behaviors of carbon vacancy under external elements adsorption provide a very valuable method to manipulate the magnetism of Graphene. Proximity to ferromagnetic materials like Ni can also induce magnetism in Graphene. In case of Ferromagnetic insulators in contact with Graphene, atomic orbital overlap at the interface induces a spin-splitting in the Graphene layer and thereby induces magnetism.

Not only defects can produce magnetism, atom or molecule adsorptions also can lead to the occurrence of magnetic moments. Studies have been made on the adsorption of hydrogen atoms on Graphene. The results confirmed that such adsorption will lead to

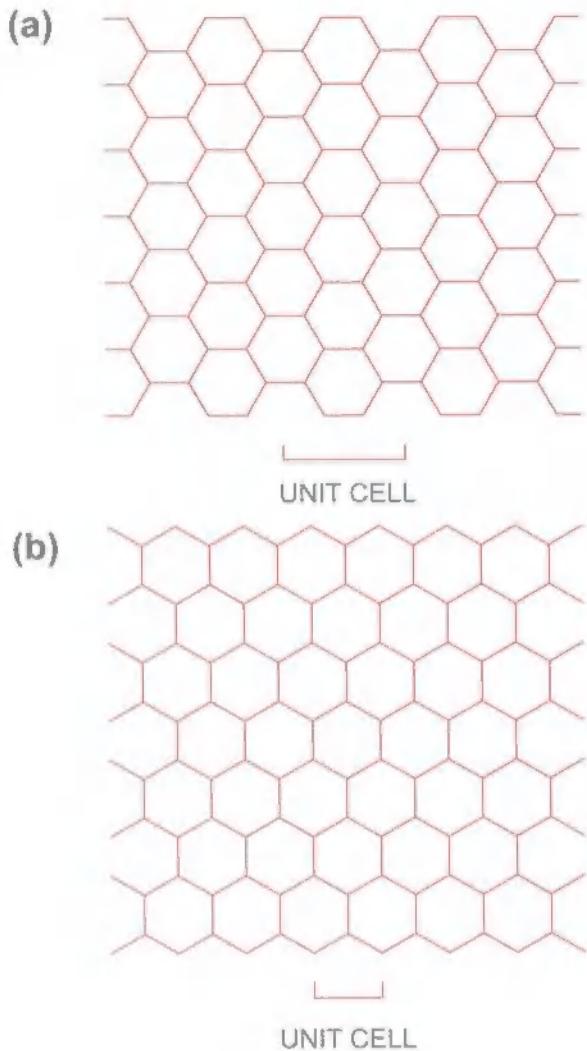


Figure 6: Armchair (a) and Zigzag (b) Edges in GNR

magnetic moments on neighboring carbon atoms, and such spin-polarized states are mainly localized around the adsorptive hydrogen. Another feature is that the sp^2 carbon atoms will become sp^3 carbon, and make the Graphene lose the D_{3h} symmetry.

The electronic structure of Graphene-based nanostructures is expected to be different from bulk Graphene because of surface, or, more properly, edge effects. This is particularly true in the case of structures with zigzag edges which present magnetic properties. Calculations confirm that these bands are prone to magnetic instabilities. Graphene Nano ribbons (GNRs) are one-dimensional stripes "cut" from Graphene, which are a few nanometers in width and quasi-infinite in the

perpendicular direction. Cutting the Graphene lattice along two different crystallographic directions, we get two kinds of edges in the system: zigzag and armchair like edges. Their electronic and magnetic properties are extremely different.

GNRs with zigzag edges (ZGNRs) are of particular interest, because they are forming edge states. The latter are localized electronic states that decay exponentially toward the center of the ribbon. The magnetic moment would depend on the total length of zigzag segments and in principle can be arbitrarily large. At first glance, this mechanism seems to lack an explanation for the value of MS being much smaller than the available broken bonds could generate. However, a recent theory suggests that, due to interactions between different zigzag segments in sub-100 nm samples of a random shape, just a small number of non-compensated spins can survive (<10), which depends on sample size only logarithmically.

In a planar sp^2 system like Graphene, the electronic states split into in-plane (σ) and out-of-plane (π) states that are decoupled by symmetry. At an unpassivated zigzag edge the hexagonal carbon network is interrupted, and both the σ and the π system form edge states. The edge states of the σ system are unpaired electrons in sp^2 orbitals, i.e., dangling σ bonds. The edge states of the π system are the ones that are of particular interest. Within the nonmagnetic, electronic band structure of an unpassivated ZGNR both sets of edge states appear as nearly flat bands (or as flat parts of bands) at E_F . Introduction of armchair edges opens a gap which varies with the ribbon width. This structure is semiconducting and no magnetic order is predicted. As a natural counterpart of armchair GNRs, carbon nanotubes (CNT) provide many valuable insights for the magnetic research. Besides the substantial magnetism, research also shows that such structures may display half-metallicity.

The spin magnetic moments in adjacent carbon atoms in a Graphene sheet are antiparallel (indicated with red and blue circles in figure 7). This means that mutual orientation for magnetic moments belonging to opposite edges is antiparallel or the edge states become spin polarized. In the magnetic ground state a band gap at E_F is opened and the atoms are ferromagnetically ordered along one edge and anti-ferromagnetically ordered between opposite edges. Structures like the six-edge hexagon give high expectation value of the spin. As the density of states is localized at the edges, the

magnetic moment decreases while moving towards the center of the ribbon. The overall magnetic moment decreases with increasing number of armchair edges introduced along the non-horizontal directions. The increment in the number of armchair edges corresponds in shortening zigzag ones.

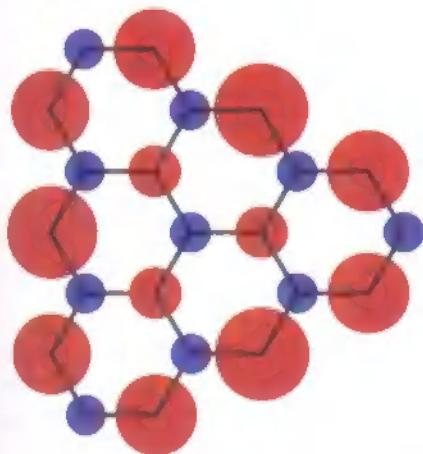


Figure 7: Direction of spin magnetic moment in Graphene (red is up and blue is down)

Because the magnetic states are the edge states, the external transverse electric fields are expected to have significant effect on such states. When an external electric field is applied across the ribbons, the applied electric fields remove the degeneracy in energy of the two edges, and make the ribbons spin selective. The break of degeneracy is due to the additional potential caused by the electric fields. Electric-field control of magnetic properties has been subject of considerable attention, with a few materials and hybrid magnetic systems found recently where it is possible to control the magnetization direction and/or the Curie temperature. Electric field tunability is one of the fundamental properties of Graphene and has been widely used to control its electronic, optical and other properties related to the electronic structure. On the other hand, Graphene is believed to be an ideal material for spintronics due to the weak spin-orbit interaction and long spin relaxation lengths and the possibility to introduce paramagnetic centers via controlled introduction of defects, as recent experiments have shown that both vacancies and adatoms in Graphene carry magnetic moments $\mu \approx \mu_0$. In principle, the latter is not exceptional because defects and impurities in crystals without any d or f elements may have unpaired electrons and, therefore, exhibit paramagnetism.

However, achieving ferromagnetic alignment between these unpaired spins has proven practically impossible. In Graphene the prospect of defect ferromagnetism is more realistic because, firstly, the presence of conduction electrons provides a medium for coupling between localized spins and, secondly, the defect-induced moments are believed to be due to the same π electrons that are responsible for electron transport. Accordingly, it should be possible to use the electric-field effect to control not only the coupling between localized magnetic moments in Graphene, but also the presence of the moments themselves which so far has not been possible in other materials.

Potential applications of Graphene

Graphene and its derivatives have shown outstanding potentials in many fields such as nano-electronics, nano-composite materials, energy storage, field effect transistor (FET), organic light emission diodes (OLED), sensors and biomedical applications. We are however restricting our discussion by mentioning a few of its electronic applications.

Because of high electrical conductivity, mechanical flexibility and low cost, Graphene and its derivatives have got wide spread applications in light emitting diode (LED), field effect transistor (FET), memory and photovoltaic devices. Due to unique band structure, the carriers in Graphene are bipolar, with electrons and holes that can be continuously tuned by a gate electrical field. Graphene based FET is possible using Graphene Nano Ribbons (GNR) which has a suitable band gap for FET applications. Though Graphene based MOSFET may not be practically useful, it is not required either, as Graphene FETs have much faster switching than Si based MOSFETs.

With high electrical conductivity, high carrier mobility and moderately high optical transmittance in the visible range of spectrum, Graphene show promise for Transparent Conductive Films (TCFs) and is a strong contender for future optoelectronic devices including solar cells. Research has also shown that the performance of Li battery can be substantially enhanced using $\text{SnO}_2/\text{Graphene}$ or $\text{SnSb}/\text{Graphene}$ hybrid structures. Graphene is also a potential candidate for various kinds of high efficiency sensors as theoretically it can detect single molecules; thus it may open up the new area of molecular electronics.

Another area of much interest is Graphene's potential use in Spintronics. The concept of Spintronics is based on transfer of spin instead of charged carriers by ferromagnetic materials as the main transport phenomenon. Since Graphene shows magnetism and has a spin polarized electronic structure, it is being considered as the most potential candidate for spintronics application, which may herald a post-Silicon era in electronics and revolutionize it.

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